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<b>(54) Title:</b> IMPACT-MODIFIED POLYMER BLENDS CONTAINING A LIQUID CRYSTALLINE POLYMER  <b>(57) Abstract</b>  This invention relates to a blend comprising: (a) about 50 to about 98 weight percent of an isotropic polymer containing at least one epoxy functional monomer having at least one epoxy group, and (b) about 2 to about 50 weight percent of a liquid crystalline polyester.		

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**IMPACT-MODIFIED POLYMER BLENDS CONTAINING  
A LIQUID CRYSTALLINE POLYMER**

Field of the Invention

5        This invention relates to polymer blends in which a liquid crystalline polymer is blended with a polymer which contains an epoxy functionality.

Background of the Invention

10       The blending of liquid crystalline polymers with isotropic polymers to obtain mechanical property improvements is known.

      For example, a blend of 85-95 weight % wholly aromatic liquid crystalline polyester (LCP) with 5-15  
15       weight % of a polyalkylene terephthalate which resulted in improved mechanical properties is described in United States Patent 4,451,611.

      Similarly, blending 5-75 weight % polycarbonate with 25-95 weight % of a wholly aromatic liquid  
20       crystalline polyester resulted in improved tensile and flexural properties as described in United States Patent 4,460,735.

      In the prior art, addition of LCP's to isotropic polymers often produces materials with improved  
25       strength, stiffness and processability, but generally at the expense of decreasing impact strength with increasing LCP concentration.

      In the present invention, a small amount of liquid crystalline polyester is added to modify the properties  
30       of a non-polyester matrix polymer. Surprisingly, the incorporation of epoxy functionality into the matrix polymer prevents the concomitant decrease in its impact strength normally associated with incorporation of the LCP.

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There are no known patents which describe the addition of a liquid crystalline polyester to a polymer containing a reactive group, though blends of epoxy-modified polymers with other polyesters have been reported. The use of epoxy-functional polymers in blends with polyesters such as polyethylene terephthalate and polybutylene terephthalate is disclosed, for example, in United States Patent 4,284,540. This patent describes the addition of a minor amount of epoxy-modified polymer to the polyester matrix to modify the properties of the polyester.

Additional blends of polyesters with small amounts of epoxy-modified polymers are disclosed in United States Patent 5,206,291. In the prior art, it is generally found that a minor amount of an appropriate epoxy-modified polymer can be effective at raising the impact strength of the polyester composition and that this increase in impact strength is accompanied by an undesired decrease in modulus.

In the compositions of the present invention, a minor amount of a liquid crystalline polyester is added to an isotropic polymer matrix containing epoxy-functionality. A surprising balance of impact strength and modulus is obtained with blends of this invention.

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#### Summary of the Invention

This invention relates to a class of such blends in which the isotropic polymer contains a reactive epoxy group.

More particularly, this invention relates to a blend comprising:

- (a) about 50 to about 98 weight percent of an isotropic polymer containing at least one epoxy functional monomer having at least one epoxy group, and

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(b) about 2 to about 50 weight percent of a liquid crystalline polyester.

This invention also relates to a blend comprising:

- 5 (a) about 2 to about 30 weight percent of an isotropic polymer containing at least one epoxy functional monomer having at least one epoxy group,
- (b) about 2 to about 50 weight percent of a liquid crystalline polyester, and
- 10 (c) about 96 to about 20 weight percent of an isotropic polymer which does not contain an epoxy group.

15 This invention unexpectedly results in properties which are superior to those of the isotropic polymer, including an impact strength which is unexpectedly higher than that of the isotropic polymer over a wide temperature range.

20 Detailed Description of the Preferred Embodiments

This invention relates to a blend comprising:

- (a) about 50 to about 98, preferably about 70 to about 95, weight percent of an isotropic polymer containing at least one epoxy-
- 25 functional monomer having at least one epoxy group, and
- (b) about 2 to about 50, preferably about 5 to about 30, weight percent of a liquid crystalline polyester.

30 The blend of this invention may optionally contain an isotropic polymer (C) which does not contain an epoxy-functional monomer containing an epoxy group. In this instance, the blend would comprise

- (a) about 2 to about 30, preferably about 5 to
- 35 about 20, weight percent of an isotropic

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polymer containing at least one epoxy-functional monomer having at least one epoxy group,

5 (b) about 2 to about 50, preferably about 5 to about 30, more preferably about 10 to about 20 weight percent, of a liquid crystalline polyester and

10 (c) about 96 to about 20, preferably about 90 to about 50, more preferably about 85 to about 60 weight percent of an isotropic polymer which does not contain an epoxy group.

The functionality of the epoxy functional polymer of the invention should be such that the average number of epoxy groups per polymer chain, based on the number  
15 average molecular weight of the polymer, is between about 1 to about 100, and more preferably, between about 6 and 75.

Epoxy functional comonomers useful in preparing the epoxy functional polymers of the invention are  
20 preferably present in an amount of about 0.5 to 10, preferably about 0.5 to 5, and more preferably 1-3 mole %.

The epoxy functional polymers may be produced by any of the polymerization techniques known in the art.  
25 A particularly useful technique is free radical copolymerization of monomers containing both vinyl and epoxide functionality such as glycidyl methacrylate (GMA). It is also possible to use epoxy functional polymers containing more than one other monomer and/or  
30 functional group.

In principle, any radically copolymerizable structure containing an epoxy (oxirane) group could be incorporated in the functional blend copolymer component. The majority of the glycidyl-functional  
35 monomers are prepared by the condensation reactions of

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epichlorohydrin with hydroxy-functional vinyl compounds.

The isotropic polymer preferably may be prepared by the copolymerization of one or more monomers having vinyl and epoxy functionality selected from the group consisting of glycidyl acrylate, glycidyl methacrylate, hydroxy-alkyl acrylates, methacrylates, vinyl benzyl glycidyl ethers and epoxy butene.

In general, the monomers containing both vinyl and epoxide functionality can be copolymerized with other vinyl monomers free radically. Typical vinyl monomers without epoxide functionality which can be copolymerized with the vinyl epoxides include acrylic acid, methacrylic acid, acrylates, methacrylates, vinyl acetate, acrylonitrile, methacrylonitrile, styrene, substituted styrenes, olefins, 1,1-alkyl olefins, and the like.

The vinyl monomers without epoxide functionality are preferably present in an amount of 99.5 to 95, preferably 99-97 mole %.

A preferred isotropic polymer is an ethylene/glycidyl methacrylate copolymer, preferably comprising 1.2 mole % glycidyl methacrylate or 2.6 mole % glycidyl methacrylate.

The preferred molecular weight of the epoxy functional polymer will depend upon the nature of the copolymer. Addition polymers typically have Mw's in the range from about 50,000 to about 500,000.

Alternatively, epoxy groups could be grafted onto the non-copolymerized isotropic polymer.

The liquid crystalline polymer of the invention is a melt processable partially aromatic or wholly aromatic copolyester which is capable of forming, apart from the blend, an optical anisotropic melt phase preferably at temperatures below 325°C. Liquid crystalline polyesters suitable for the present invention commonly exhibit a

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weight average molecular weight of about 5,000 to 2,000,000 and preferably about 10,000 to 100,000. Such molecular weight may be determined by standard techniques not involving the solutioning of the polymer, e.g., by end group determination via infrared spectroscopy on compression molded films. Alternatively, light scattering techniques in a pentafluorophenol solution may be employed to determine the molecular weight.

For the purposes of the present invention, the rings which are included in the polymer backbones of the polymer components commonly include substitution of at least some of the hydrogen atoms present upon a ring. Such substituents include alkyl groups of up to five carbon atoms; alkoxy groups having up to five carbon atoms; halogens; and additional aromatic rings, such as phenyl and substituted phenyl groups. Halogens which may be listed as possible substituents include fluorine, chlorine, and bromine. Although bromine atoms tend to be released from organic compounds at high temperatures, bromine is more stable on aromatic rings than on aliphatic chains, and therefore is suitable for inclusion as a possible substituent on the rings.

The above-described polyesters, in order to be useful in the blend of the present invention, must exhibit optical anisotropy in the melt phase. These polyesters readily form liquid crystals in the melt phase and accordingly exhibit a high tendency for the polymer chains to orient in the shear direction. Such anisotropic properties are manifested at a temperature at which the wholly aromatic polyesters readily undergo melt processing to form shaped articles. The anisotropic properties may be confirmed by the use of a Leitz polarizing microscope at a magnification of 40X with the sample on a Koffler hot stage and under a



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nitrogen atmosphere. The melt phases of the wholly aromatic polyesters which are suitable for use in the present invention are optically anisotropic, i.e., they transmit light when examined between crossed-polarizers. By contrast, the melt of a conventional polymer will not appreciably transmit light when placed between crossed-polarizers.

The wholly aromatic polyesters described above are useful as molding resins and may also be used in the formation of coatings, fibers, and films. They may be molded by injection molding and can be processed by any melt extrusion technique.

The wholly aromatic polyesters consist of at least two recurring moieties each of which contributes at least one aromatic ring to the polymer backbone after being combined in the polyester.

Wholly aromatic polyester resins long have been known. For instance, 4-hydroxybenzoic acid homopolymer and copolymers have been provided in the past and are commercially available. Certain of the wholly aromatic polyesters encountered in the prior art tend to be somewhat intractable in nature and to present substantial difficulties if one attempts to melt process the same while employing conventional melt processing techniques. Such polymers are typically crystalline in nature, relatively high melting or possess a decomposition temperature which is below the melting point, and when molten, frequently exhibit an isotropic melt phase. Molding techniques such as compression molding or sintering may be utilized with such materials; however, injection molding, melt spinning, etc., commonly have not been viable alternatives or when attempted commonly have been accomplished with difficulty.

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It is to be understood that the wholly aromatic polyesters which are suitable for use in the present invention are limited to those which are capable of undergoing melt phase processing, i.e., those which exhibit no substantial decomposition at or below the melting temperature.

The wholly aromatic polyesters which are suitable for use in the present invention may be formed by a variety of ester-forming techniques whereby organic monomer compounds possessing functional groups which upon condensation form the requisite recurring moieties are reacted. For instance, the functional groups of the organic monomer compounds may be carboxylic acid groups, hydroxyl groups, ester groups, acyloxy groups, acid halides, etc.

The organic monomer compounds may be reacted in the absence of a heat exchange fluid via a melt acidolysis procedure. Also, they may be heated initially to form a melt solution of the reactants with the reaction continuing as solid polymer particles are suspended therein. A vacuum may be applied to facilitate removal of volatiles formed during the final stage of the condensation (e.g., acetic acid or water).

Also, a slurry polymerization process may be employed as shown in United States Patent 4,083,829, incorporated by reference in its entirety. In such a process, the solid product is suspended in a heat exchange medium.

When employing either the melt acidolysis procedure of the slurry procedure, the organic monomer reactants from which the wholly aromatic polyesters are derived may be initially provided in a modified form whereby the usual hydroxy groups of such monomers are esterified (i.e., they are provided as lower acyl esters). The lower acyl groups preferably have about two to about

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four carbon atoms. Preferably, the acetate esters of organic monomer reactants are provided.

Representative catalysts which optionally may be employed in either the melt acidolysis procedure or in the slurry procedure described above include dialkyl tin oxide (e.g., dibutyl tin oxide), diaryl tin oxide, titanium alkoxides, alkali and alkaline earth metal salts of carboxylic acids (e.g., zinc acetate), the gaseous acid catalysts, such as Lewis acids (e.g.,  $\text{BF}_3$ ), hydrogen halides (e.g.,  $\text{HCl}$ ), etc. The quantity of catalyst utilized typically is about 0.001 to about 1 percent by weight based upon the total monomer weight, and most commonly about 0.01 to 0.2 percent by weight.

The wholly aromatic polyesters suitable for use in the present invention tend to be substantially insoluble in common polyester solvents and accordingly are not susceptible to solution processing. As discussed previously, they can be readily processed by common melt processing techniques. Most suitable wholly aromatic polyesters are soluble in pentafluorophenol/1,2,4-trichlorobenzene mixtures.

Examples of preferred wholly aromatic polyesters suitable for the present invention are disclosed in U.S. Pat. Nos. 4,067,852; 4,083,829; 4,130,545; 4,161,470; 4,184,996; 4,238,559; 4,238,598; and 4,256,624.

A preferred liquid crystalline polyester (A) of the invention comprises repeating units from:

- (a) 2,6-naphthalenedicarboxylic acid,
- (b) 4,4'-biphenol,
- (c) hydroquinone, and
- (d) p-hydroxybenzoic acid,

wherein the molar ratios of (b) + (c) = (a) and the molar ratios of (a) + (d) = 1.0.

Another preferred liquid crystalline polyester (B) of the invention comprises repeating units from:

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- (a) 2,6-naphthalenedicarboxylic acid,
- (b) terephthalic acid,
- (c) hydroquinone, and
- (d) p-hydroxybenzoic acid,

5 wherein molar ratios of (a) + (b) = (c) and the molar ratios of (c) + (d) = 1.0.

The mole percentages of (a) - (d) for Polyester (B) are preferably as follows:

- (a) about 10 to about 30 mole %,
- 10 (b) about 10 to about 30 mole %,
- (c) about 30 to about 60 mole %,
- (d) about 40 to about 80 mole %.

Preferably, for Polyester (B), the mole percentage of (a) is about 20 mole %, the mole percentage of (b) is  
15 about 20 mole %, the mole percentage of (c) is about 40 mole % and the mole percentage of (d) is about 60 mole %.

The preferred liquid crystalline polyesters of the invention typically have an inherent viscosity of at  
20 least 2, and preferably 2 to 10, as determined in a 60/40 weight/weight solution of pentafluorophenol/1,2,4-trichlorobenzene at 25°C at a concentration of 0.1 g polyester per 100 ml solvent.

A preferred wholly aromatic copolyester consists  
25 of about 70 mole % p-hydroxybenzoic acid and about 30 mole % 2,6-hydroxynaphthoic acid.

The partially aromatic copolyesters useful in the invention typically consist of aromatic dicarboxylic acids combined with aliphatic glycols. Examples of  
30 useful aromatic dicarboxylic acids include terephthalic acid, isophthalic acid, 2-chloroterephthalic acid, 2-methylterephthalic acid, 2,5-dichloroterephthalic acid, 1,3-, 1,4-, 1,5-, 1,6-, 2,6-, or 2,7-naphthalene-dicarboxylic acid, trans-4,4'-stilbenedicarboxylic acid  
35 and 4,4'-biphenyldicarboxylic acid.

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Examples of aliphatic glycols useful in preparing the partially aromatic copolyesters of the invention include ethylene glycol, 1,6 hexanediol, 1,2-propylene glycol, 1,3-propanediol, 2,2-dimethyl-1,3-propanediol, 5 1,4-butanediol, 1,5-pentanediol, 1,3- or 1,4-cyclohexane-dimethanol(cis or trans), 2,2,4,4-tetramethyl-1,3-cyclobutanediol, and p-xylenediol.

Other examples of partially aromatic polyesters suitable for the present invention are disclosed in U.S. 10 Pat. Nos. 3,778,410; 3,804,805; 3,842,040; 4,082,731; 4,459,402; and 4,973,654, all of which are incorporated herein by reference in their entirety.

Furthermore, additional components such as stabilizers, fillers, flame retardants, colorants, 15 additional polymers, impact modifiers, nucleating agents, mold release agents, additional catalysts, and other additives known to those skilled in the art may be present during the melt blending step or added subsequently.

20 In addition, hydroxybenzoic acids or a small amount of aliphatic dicarboxylic acids may be incorporated into the polyester.

A preferred partially aromatic copolyester is produced by modifying poly(ethylene terephthalate) with 25 about 60 mole % p-hydroxybenzoic acid.

The present invention is also directed to molded objects regardless of the specific methodology employed for their preparation. Injection molding is particularly preferred, however.

30 The blends are compounded in the melt, for example by using a single screw or twin screw extruder. Furthermore, additional components such as stabilizers, fillers, flame retardants, colorants, additional polymers, impact modifiers, nucleating agents, mold 35 release agents, additional catalysts, and other

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additives known to those skilled in the art may be present during the melt blending step or added subsequently.

5 This invention can be further illustrated by the following examples of preferred embodiments thereof, although it will be understood that these examples are included merely for purposes of illustration and are not intended to limit the scope of the invention unless otherwise specifically indicated. All percentages are  
10 by weight, unless otherwise specified.

#### EXAMPLES

15 The invention is illustrated for the following liquid crystalline polymers:

LCP A: Vectra A950, a product of Hoechst-Celanese. A wholly aromatic copolyester consisting of 70 mole percent p-hydroxybenzoic acid and 30 mole percent 2,6-  
20 hydroxynapthoic acid.

LCP B: Rodrun 3000, a product of the Unitika Chemical Company. A partially aromatic copolyester produced by modifying poly(ethylene terephthalate) with 60 mole  
25 percent p-hydroxybenzoic acid.

The following isotropic polymers were used.

EGMA 2C: Igetabond 2C, a product of Sumitomo Chemical  
30 Co. Ethylene/glycidyl methacrylate copolymer containing 1.2 mole % glycidyl methacrylate (GMA). Weight average molecular weight is 123,000;  $M_w/M_n = 6.0$ . Melt index = 3.0 g/10 min. Approximately 10 GMA groups per chain.  
(Invention)

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EGMA E: Igetabond E from Sumitomo, 2.6 mole % GMA.  $M_w = 123,000$ ;  $M_w/M_n = 5.6$ . Melt index = 3.0 g/10 min. Approximately 20 GMA groups per chain. (Invention)

5 EVA: Elvax 760, E.I. DuPont. Vinyl acetate concentration 9.3 wt % (3.3 mole%); Melt index = 2.0 g/10 min. (Comparative)

10 PE: Tenite 1550 low density polyethylene. (Comparative)

Example 1:

EGMA E was blended with 10 and 20 weight percent of LCP A. The blends were compounded at 270°C and injection  
15 molded at 270°C. The data are shown in Table 1. Incorporation of the LCP causes an unexpected increase in the impact strength of the blends at all three measurement temperatures.

20 Example 2: (Comparative)

For comparison purposes, both PE and EVA were blended with the higher concentration of 20 weight percent of LCP A. EVA is a modified polyethylene, but it does not contain epoxy modification. PE is an unmodified  
25 polyethylene. Samples were compounded at 270°C and injection molded at 270°C. The data are shown in Table 2. At the two higher measurement temperatures, addition of the LCP causes a significant decrease in the impact strength of the unmodified polyethylene. The  
30 impact strength of the EVA blend remains constant at 23°C and a small decrease is observed at 0°C. At the lowest measurement temperature of -40°C, addition of the LCP causes a very large decrease in impact strength for both of these polyethylenes. These results demonstrate  
35 that it is the epoxy functionality of the modified

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polyolefin in Example 1 which yields the unexpected results.

Example 3:

5 EGMA 2C was blended with 15 and 30 weight percent of LCP  
B. The blends were compounded at 200°C and injection  
molded at 270°C. The data are shown in Table 3.  
Incorporation of the LCP causes a significant increase  
in the flexural modulus of the EGMA 2C. Concomitantly  
10 there is also an unexpected increase in the impact  
strength of the blends.

Example 4: (Comparative)

15 PE was blended with 30 weight percent of LCP B. This  
blend was also compounded at 200°C and injection molded  
at 270°C. The data are shown in Table 4. Addition of  
the LCP causes an increase in flexural modulus which is  
comparable to that observed in Example 3. However, the  
blend has an impact strength which is significantly  
20 reduced from that of the pure polyethylene.

Example 5:

In this example, PE was pre-blended with 6 and 21 weight  
percent of EGMA E. The 6% pre-blend was then compounded  
25 with 15 weight percent of LCP B and the 21% pre-blend  
was compounded with 30 weight percent of LCP B. The  
data are shown in Table 5. Addition of the LCP to these  
PE-EGMA E blends results in an increase in impact  
strength over that measured for the pure polyethylene.  
30 The data shown previously in Table 4 indicated that the  
addition of LCP B to unmodified PE results in a large  
reduction in impact strength. Comparison of those  
results to the present example demonstrates that it is  
the pre-blending with EGMA-E which causes the observed  
35 increase in impact strength upon addition of LCP B.



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Table 1. The effect of LCP A on the impact strength of EGMA E.

<u>%LCP A</u>		<u>0.</u>	<u>10.</u>	<u>20.</u>
5	Unnotched Izod, 23°C	4.5	5.8	6.1
	ft-lb/in      0°C	8.3	9.3	11.6
	-40°C	23.5	26.3	31.7

10

Table 2. The effect of LCP A on the impact strength of PE and EVA.

<u>%LCP A</u>		<u>PE</u>		<u>EVA</u>	
		<u>0.</u>	<u>20.</u>	<u>0.</u>	<u>20.</u>
15	Unnotched Izod, 23°C	9.2	4.3	5.8	5.9
	ft-lb/in      0°C	12.3	3.5	9.0	5.1
	-40°C	25.4	3.3	23.4	3.6

20

Table 3. The effect of LCP B on the impact strength and flexural modulus of EGMA 2C.

<u>%LCP B</u>		<u>0.</u>	<u>15.</u>	<u>30.</u>
25	Unnotched Izod, 23°C	7.5	10.1	13.4
	ft-lb/in			
	Flexural modulus	0.10	0.27	1.00
	10 <sup>5</sup> PSI			

30

Table 4. The effect of LCP B on the impact strength and flexural modulus of PE.

<u>%LCP B</u>		<u>0.</u>	<u>30.</u>
35	Unnotched Izod, 23°C	8.0	2.8
	ft-lb/in		
40	Flexural modulus,	0.12	1.41
	10 <sup>5</sup> PSI		

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Table 5. The effect of LCP B on the impact strength of PE/EGMA E blends.

5	% LCP B Polyolefin	0	15	30
		<u>PE</u>	<u>PE(6%EGMA E)</u>	<u>PE(21% EGMAE)</u>
	Unnotched Izod, 23°C	8.8	11.2	13.1
	ft-lb/in            0°C	13.7	16.4	18.6

10

The invention has been described in detail with particular reference to preferred embodiments thereof, but it will be understood that variations and modifications can be effected within the spirit and scope of the invention. Moreover, all patents, patent applications (published or unpublished, foreign or domestic), literature references or other publications noted above are incorporated herein by reference for any disclosure pertinent to the practice of this invention.

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CLAIMSWe claim:

1. A blend comprising:
  - (a) about 50 to about 98 weight percent of an isotropic polymer containing at least one epoxy functional monomer having at least one epoxy group, and
  - (b) about 2 to about 50 weight percent of a liquid crystalline polyester.
2. The blend of Claim 1 wherein said liquid crystalline polyester is present in an amount of 5-30 weight %.
3. The blend of Claim 1 wherein the average number of said epoxy groups per polymer chain is from about 1 and about 100.
4. The blend of Claim 3 wherein the average number of said epoxy groups per polymer chain is from about 6 to about 75.
5. The blend of Claim 1 wherein said epoxy functional monomer is present in an amount of about 0.5 to 5.0 mole percent.
6. The blend of Claim 5 wherein said epoxy functional monomer is present in an amount of about 1.0 to 3.0 mole percent.
7. The blend of Claim 1 wherein said isotropic polymer is an addition polymer.

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8. The blend of Claim 7 wherein said addition polymer has a weight average molecular weight of from about 50,000 to about 500,000.
- 5 9. The blend of Claim 1 wherein said isotropic polymer containing at least one epoxy functional monomer comprises:
- 10 (a) one or more vinyl monomers without epoxide functionality selected from the group consisting of acrylic acid, methacrylic acid, acrylates, methacrylates, vinyl acetate, acrylonitrile, methacrylonitrile, styrene, substituted styrenes, olefins, and 1,1-alkyl olefins, and
- 15 (b) one or more monomers having both vinyl and epoxide functionality selected from the group consisting of glycidyl acrylate, glycidyl methacrylate, hydroxy-alkyl acrylates, methacrylates, vinyl benzyl glycidyl ethers
- 20 and epoxy butene.
10. The blend of Claim 9 wherein the mole percent of (a) is about 99.5 to 95.
- 25 11. The blend of Claim 9 wherein the mole percent of (a) is about 99.0 to 97.0.
12. The blend of Claim 9 wherein said isotropic polymer is an ethylene/glycidyl methacrylate copolymer.
- 30 13. The blend of Claim 12 wherein said ethylene/glycidyl methacrylate copolymer comprises 1.2 mole % glycidyl methacrylate.
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14. The blend of Claim 12 wherein said ethylene/glycidyl methacrylate copolymer comprises 2.6 mole % glycidyl methacrylate.
- 5 15. The blend of Claim 1 wherein said liquid crystalline polyester is a wholly aromatic copolyester.
- 10 16. The blend of Claim 15 wherein said liquid crystalline polyester has an inherent viscosity of at least 2 as determined in a 60/40 weight/weight solution of pentafluorophenol/1,2,4-trichlorobenzene at 25°C at a concentration of 0.1 g polyester per 100 ml solvent.
- 15 17. The blend of Claim 16 wherein said liquid crystalline polyester has an inherent viscosity of 2 to 10 as determined in a 60/40 weight/weight solution of pentafluorophenol/1,2,4-trichlorobenzene at 25°C at a concentration of 0.1 g polyester per 100 ml solvent.
- 20 18. The blend of Claim 15 wherein said liquid crystalline polyester comprises repeat units from:
- 25 (a) 2,6-naphthalenedicarboxylic acid,  
(b) 4,4'-biphenol,  
(c) hydroquinone, and  
(d) p-hydroxybenzoic acid,  
wherein the molar ratios of (b) + (c) = (a) and  
30 the molar ratios of (a) + (d) = 1.0.
19. The blend of Claim 15 wherein said liquid crystalline polyester comprises repeat units from:
- 35 (a) 2,6-naphthalenedicarboxylic acid,  
(b) terephthalic acid,

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(c) hydroquinone, and

(d) p-hydroxybenzoic acid

wherein the molar ratios of (a) + (b) = (c) and  
the molar ratios of (c) + (d) = 1.0.

5

20. The blend of Claim 19 wherein the mole percentages  
of (a) - (d) comprise the following:

(a) about 10 to about 30 mole %,

(b) about 10 to about 30 mole %,

10

(c) about 30 to about 60 mole %, and

(d) about 40 to about 80 mole %.

21. The blend of Claim 15 wherein said wholly aromatic  
copolyester consists of about 70 mole % p-  
15 hydroxybenzoic acid and about 30 mole % 2,6-  
hydroxynaphthoic acid.

22. A molded object prepared using the blend of  
Claim 15.

20

23. The blend of Claim 1 wherein said liquid  
crystalline polyester is a partially aromatic  
copolyester.

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24. The blend of Claim 23 wherein said partially  
aromatic copolyester is produced by modifying  
poly(ethylene terephthalate) with about 60 mole %  
p-hydroxybenzoic acid.

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25. A blend comprising:

(a) about 2 to about 30 weight percent of an  
isotropic polymer containing at least one  
epoxy functional monomer having at least one  
epoxy group,

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- (b) about 2 to about 50 weight percent of a liquid crystalline polyester, and  
(c) about 96 to about 20 weight percent of an isotropic polymer which does not contain an epoxy group.
- 5
26. The blend of Claim 25 wherein (a) is present in an amount of about 5 to about 20 weight percent, (b) is present in an amount of about 5 to about 30 weight percent, and (c) is present in an amount of about 90 to about 50 weight percent.
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27. The blend of Claim 25 wherein said liquid crystalline polyester is present in an amount of 10-20 weight %.
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28. The blend of Claim 25 wherein the average number of said epoxy groups per polymer chain is from about 1 and about 100.
- 20
29. The blend of Claim 28 wherein the average number of said epoxy groups per polymer chain is from about 6 to about 75.
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30. The blend of Claim 25 wherein said epoxy functional monomer is present in an amount of about 0.5 to 5.0 mole percent.
31. The blend of Claim 30 wherein said epoxy functional polymer is present in an amount of about 1.0 to 3.0 mole percent.
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32. The blend of Claim 25 wherein said isotropic polymer is an addition polymer.
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33. The blend of Claim 25 wherein said addition polymer has a weight average molecular weight of from about 50,000 to about 500,000.
- 5 34. The blend of Claim 25 wherein said isotropic polymer containing at least one epoxy functional monomer comprises:
- 10 (a) one or more vinyl monomers without epoxide functionality selected from the group consisting of acrylic acid, methacrylic acid, acrylates, methacrylates, vinyl acetate, acrylonitrile, methacrylonitrile, styrene, substituted styrenes, olefins, and 1,1-alkyl olefins, and
- 15 (b) one or more monomers having both vinyl and epoxide functionality selected from the group consisting of glycidyl acrylate, glycidyl methacrylate, hydroxy-alkyl acrylates, methacrylates, vinyl benzyl glycidyl ethers
- 20 and epoxy butene.
35. The blend of Claim 34 wherein the mole percent of (a) is about 99.5 to 95.
- 25 36. The blend of Claim 35 wherein the mole percent of (a) is about 99.0 to 97.0.
37. The blend of Claim 34 wherein said isotropic polymer is an ethylene/glycidyl methacrylate copolymer.
- 30 38. The blend of Claim 37 wherein said ethylene/glycidyl methacrylate copolymer comprises 1.2 mole % glycidyl methacrylate.

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39. The blend of Claim 37 wherein said ethylene/glycidyl methacrylate copolymer comprises 2.6 mole % glycidyl methacrylate.
- 5 40. The blend of Claim 25 wherein said liquid crystalline polyester is a wholly aromatic copolyester.
- 10 41. The blend of Claim 40 wherein said liquid crystalline polyester has an inherent viscosity of at least 2 as determined in a 60/40 weight/weight solution of pentafluorophenol/1,2,4-trichlorobenzene at 25°C at a concentration of 0.1 g polyester per 100 ml solvent.
- 15 42. The blend of Claim 41 wherein said liquid crystalline polyester has an inherent viscosity of 2 to 10 as determined in a 60/40 weight/weight solution of pentafluorophenol/1,2,4-trichlorobenzene at 25°C at a concentration of 0.1 g polyester per 100 ml solvent.
- 20 43. The blend of Claim 40 wherein said liquid crystalline polyester comprises repeat units from:
- 25 (a) 2,6-naphthalenedicarboxylic acid,  
(b) 4,4'-biphenol,  
(c) hydroquinone, and  
(d) p-hydroxybenzoic acid,  
wherein the molar ratios of (b) + (c) = (a) and  
30 the molar ratios of (a) + (d) = 1.0.
44. The blend of Claim 40 wherein said liquid crystalline polyester comprises repeat units from:
- 35 (a) 2,6-naphthalenedicarboxylic acid,  
(b) terephthalic acid,

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(c) hydroquinone, and

(d) p-hydroxy benzoic acid

wherein the molar ratios of (a) + (b) = (c) and  
the molar ratios of (c) + (d) = 1.0.

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45. The blend of Claim 44 wherein the mole percentages  
of (a) - (d) comprise the following:

(a) about 10 to about 30 mole %,

(b) about 10 to about 30 mole %,

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(c) about 30 to about 60 mole %, and

(d) about 40 to about 80 mole %.

46. The blend of Claim 40 wherein said wholly aromatic  
copolyester consists of about 70 mole % p-  
hydroxybenzoic acid and about 30 mole % 2,6-  
hydroxynaphthoic acid.

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47. A molded object prepared using the blend of  
Claim 40.

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48. The blend of Claim 25 wherein said liquid  
crystalline polyester is a partially aromatic  
copolyester.

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49. The blend of Claim 48 wherein said partially  
aromatic copolyester is produced by modifying  
poly(ethylene terephthalate) with about 60 mole %  
p-hydroxybenzoic acid.

## INTERNATIONAL SEARCH REPORT

International application No.

PCT/US 94/13126

## A. CLASSIFICATION OF SUBJECT MATTER

IPC6: C08L 67/02, C08L 101/06

According to International Patent Classification (IPC) or to both national classification and IPC

## B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC6: C08L

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

WPI, CLAIMS, JAPIO

## C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	EPOQUE, JPO & Japio, JP5171016, NIPPON G II PLAST KK: "RESIN COMPOSITION CONTAINING LIQUID- CRYSTALLINE POLYESTER", 930709	1-8, 15-33, 40-49
Y	--	9-14, 34-39
Y	US, A, 5206291 (M. E. STEWART), 27 April 1993 (27.04.93), see column 2, line 45-column 3, line 28	9-14, 34-39
A	US, A, 4083829 (CALUNDANN ET AL), 11 April 1978 (11.04.78), see the abstract	18-21, 23, 24, 43-46, 48, 49
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☐ Further documents are listed in the continuation of Box C.
 ☒ See patent family annex.

## \* Special categories of cited documents

"A" document defining the general state of the art which is not considered to be of particular relevance

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"&amp;" document member of the same patent family

Date of the actual completion of the international search

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**INTERNATIONAL SEARCH REPORT**

Information on patent family members

SA 100676

International application No.

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			JP-T-	6504560	26/05/94
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